The beginning of quantum nonlinear optics [Invited]

Amnon Yariv

Department of Electrical Engineering and Applied Physics, California Institute of Technology, 1200 E. California Blvd. 128-95, Pasadena, California 91125, USA (email: ayariv@caltech.edu)

Received October 5, 2011; accepted October 7, 2011;
posted November 15, 2011 (Doc. ID 156099); published December 7, 2011

A description by the author of the early days of nonlinear optics (NLO), and the motivation for the formulation of the quantum theory of NLO, implications to a number of diverse areas, and the background for the prediction of spontaneous parametric fluorescence.

OCIS codes: 000.2850, 190.0190, 270.0270, 190.4975.

When asked to talk at the symposium celebrating the fiftieth birthday of nonlinear optics in Kauai last July, I needed to decide on an interesting approach beyond that of rehashing the, by now familiar, scientific facts. The work that I will describe has to do with the quantum mechanical formalism for nonlinear optics. Although the implications of this theory are fundamental and continue to unfold, the initial motivation was much more prosaic. It had to do with deciding whether the parametric amplifier (PA) was a more sensitive amplifier than the maser/laser amplifier or not. In engineering terms, the question was, "What is the noise figure or, equivalently, the noise equivalent input power of the PA?" The type of question that a communication engineer building a microwave link would ask. The following story will describe how this simple engineering question led to the quantum formulation of nonlinear optics and the attendant prediction of new phenomena which were outside the purview of classical optics.

It begins with my graduate doctoral research at the University of California (UC) Berkeley around 1956 in Professor John Whinnery's group in the department of electrical engineering. The main topic of research in the group was traveling wave tubes, which were used as high-power wideband microwave amplifiers (they are still in use in communication satellites). As in any preamplifier of weak signals, an attribute of the traveling wave tube was its noise figure, which is defined as the signal to noise ratio at the input to the amplifier divided by that at the output. A noise figure of unity (0 dB) would imply that no noise was added to the signal in the process of amplification. Much of the work at the UC laboratory and at a few other laboratories was devoted to the understanding of the noise generation mechanisms in the traveling wave tube and to means of reducing the noise. After completing a Master of Science thesis in this field, I chose an altogether different direction for my doctoral research. It involved making and characterizing a two-level microwave maser based on spin inversion in F-centers in an MgO crystal.

After graduation in 1959, I joined the Bell Telephone Laboratories (BTL) communication department, which was headed by the legendary John Pierce. The 1958 optical maser (laser) paper by Schawlow and Townes [1] had already appeared and had the effect of starting a race to make the first optical maser. I joined Bob Collins and Gary Boyd in one of three groups at Bell Labs attempting to demonstrate the first laser (a race won in 1960 by Theodore Maiman at the Hughes Research Laboratories, but that is another story).

In the fall of 1959, I attended a small workshop on Quantum Electronics—Resonance Phenomena [2], which was held at the Shawanga Lodge in the Catskill Mountains north of New York City. The meeting was organized and chaired by Professor Charles Townes at Columbia University. The list of attendees included N. Bloembergen, R. H. Dicke, C. Kittel, R. Kompfner, I. Rowe, A. E. Siegman, and many other present and future big names in the field.

Although the lasers had yet to be demonstrated—and this would not happen until some nine months later—these distinguished scientists were already discussing operational features of this yet unborn invention. Of particular relevance to this talk were two papers: the first was by R. Serber and C. H. Townes of Columbia University and was entitled "Limits on electromagnetic amplification due to complementarity" [2]. The second paper was by H. Heffner, an electrical engineering professor at Stanford University, with the title "Parametric amplifiers and their comparison with masers" [3]. These two talks resonated with my earlier interest as a graduate student in Berkeley in the subjects of noise and amplifiers.

The paper by Serber and Townes (ST) [2] is, to this day, a model of elegance and conciseness. In my case, it also provided the main impetus for learning the mathematical tools for handling quantized electromagnetic (EM) fields and their interactions. These tools were, and are, prerequisite for treating power exchange between atoms and EM fields and even between EM fields and acoustic waves. The paper started by deriving the interaction (perturbation) Hamiltonian due to the interaction of the maser (laser) atoms with the EM field. A very quick primer on the relevant quantum mechanics at this point might be in order.

A quantum mechanical system is described completely by means of its Hamiltonian $H(\bar{p}, \bar{q})$, which is a mathematical differential operator representation of the system's (atom+field) energy. $\bar{p}$ and $\bar{q}$ are quantized field coordinates. The basic (primitive) system involved in the laser amplifier comprises an atom in an excited state and an EM mode. If the atoms and the field do not interact with each other (this can happen if the atom's dipole moment is zero), then the system will remain in its pure eigenstate, in which no energy exchange between the atom and field takes place.

When not interacting with each other, the two components of our system, the atom and the EM field, obey, separately, the time-independent Schrodinger equations:

$$H_a u_a^{(\mathrm{ex})} = E_a u_a^{(\mathrm{ex})},$$

where $u_a^{(\mathrm{ex})}$ is the wave function of the atom in its excited state and is a function of spatial coordinates. The EM mode u_n obeys the equation

$$H_m u_n = (n + 1/2)\hbar\omega_m u_n,$$

where u_n is the EM mode wavefunction and is a product of a space-dependent function and the harmonic oscillator wavefunction, the integer n is equal to the number of photons in the mode, and ω_m is the (radian) frequency of the corresponding classical oscillator. The combined atom-mode system is described by the product wave function $u_{\mathrm{total}} = u_a^{(\mathrm{ex})} u_n$ with a total energy $E_a + (n + \frac{1}{2})\hbar\omega_m$,

$$H_o u_{\mathrm{total}} = (H_a + H_m) u_{\mathrm{total}} = \left[E_a + \left(n + \frac{1}{2}\right)\hbar\omega_m\right] u_{\mathrm{total}}, \tag{1}$$

where H_o is the sum of the EM mode Hamiltonian and that of the atom. Note that H_a and u_a involve atomic coordinates only, while H_m and u_n contain EM field coordinates.

The situation described above in which the atom and the EM field do not interact is of no great interest to us. In the case of a laser, the atom and the modal EM field interact strongly. This means that the atom is aware (perturbed) of the field and vice versa. Mathematically, this is represented by the addition to the total system Hamiltonian H_o, a term H_I (I for interaction) that involves both atomic and field coordinates.

$$H_{\mathrm{system}} = H_a + H_m + H_I.$$

The immediate consequence of the inclusion of H_I in the system Hamiltonian is that the system is no longer capable of remaining in a pure eigenstate $u_a u_n$ (of H_o) and is induced to make transitions to other eigenstate of H_o. In the laser amplifier, or oscillator, the excited atom undergoes a transition to a lower state, thereby releasing an energy E_a which is picked up by the EM mode as it makes a transition from u_n to u_{n+1}, thus gaining an amount $\hbar\omega_m$ of energy (i.e., a photon). A necessary condition is a (near) conservation of energy; i.e., $\hbar\omega_m \approx E_a$. This is illustrated in Fig. 1. The increase in the EM mode energy is the basis of laser amplification (the laser oscillation requires the addition of a feedback).

The quantum noise limit here is due to the fact that a transition can occur even when initially the EM mode is unexcited, i.e., $n = 0$. Using the language of quantum mechanics [4], the rate of the process described above is proportional to the squared magnitude of the "matrix element," mathematically the integral

$$\langle u_o u_{n+1} | H_I | u_a u_n \rangle \propto \iint \mathrm{d}^3 r_m \mathrm{d}^3 r_a u_0^* u_{n+1}^* \vec{r}_a u_a u_n,$$

where $\vec{r}_a$ is the atomic (electron) coordinate and r_m is the quantized EM model field coordinate. This is the fundamental process of laser amplification and oscillation, and it is illustrated in Fig. 1. The EM field, in the case of a traveling wave,

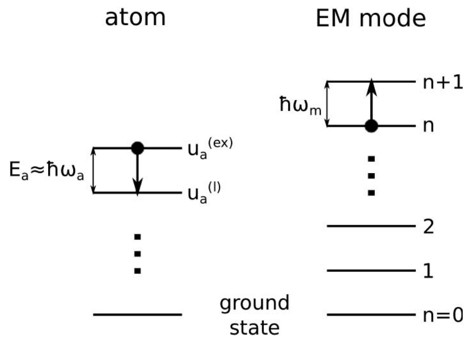

Fig. 1. The basic transition involved in the laser amplifier. The atom undergoes a transition from an excited state to a lower one while an EM mode interacting with it gains the released energy $\hbar\omega_a$.

grows coherently as it propagates in the inverted population atomic medium, inducing atoms along its path to undergo downward transitions, thus milking them of their excited state energy.

The beauty of quantum mechanics is that while the concepts just described are abstract and seemingly esoteric, their application is relatively straightforward and their predictive power almost mysterious.

The ST paper [2] predicted that a maser (laser) amplifier ideally had a noise figure of 3 dB. That corresponds to a ×2 degradation of the signal to noise power ratio between the input and the output of the amplifier. That degradation can be thought of as due to optical (or microwave) noise power which is generated within the amplifier intermingling with the signal inextricably. It can be attributed rigorously to spontaneous emission of atoms in the excited upper laser level, which takes place independently of the existing EM photons. Using this formalism ST were able to show that the laser amplifier had an equivalent noise input power of $\hbar\omega_a B$ within a (radian) bandwidth B. This translated to a minimum noise figure of 3 dB for the amplifier and implied that, in the ideal case, an amount $(G - 1)\hbar\omega_a B$ of noise power, referred to the output, was generated within the laser amplifier. The best way to view this noise generation is to refer to Fig. 1, where the process of generating an EM photon can take place even when initially the EM mode is unexcited and is in its ground state, which corresponds to the case of zero input to the amplifier.

The paper by Heffner [3] dealing with the PA used a microwave circuit model in which a strong microwave signal at a frequency ω_p ("p" for pump) was applied to modulate the capacitance or inductance in a circuit that was resonant at two lower frequencies ω_s ("s" for signal) and ω_i ("i" for idler). The modulation of the capacitor was caused by the dependence of the capacitance of a p-n junction on the applied voltage of the pump signal. An incoming signal at ω_s could be amplified by the pump signal, provided the condition $\omega_p = \omega_i + \omega_s$ was satisfied. Heffner's analysis predicted that, in principle, the PA was lossless with a noise figure approaching 0 dB under the proper loading conditions. This was in contrast to the maser (laser) amplifier analyzed by ST with its 3 dB noise figure, a limit imposed by the uncertainty principle. In the question-and-answer period that followed the presentation, I raised the question of whether the difference in the predicted noise behavior was due to the fact that the laser analysis was quantum mechanical while that of the PA was classical That question appeared in the proceedings [2].

Upon returning to the Bell Labs, I decided to try to resolve this apparent discrepancy. I realized that I needed a quantum mechanical model for the PA and took my inspiration from the laser analysis of ST [2]. I enrolled the cooperation of a friend and a colleague at the Bell Labs, Bill Louisell. Bill was a talented mathematical physicist who enjoyed working on problems related to the communication field.

A schematic description of the PA is shown in Fig. 2, which shows the pump (ω_p), signal (ω_s), and the idler (ω_i) waves exchanging power while traversing a nonlinear crystal.

A relatively strong optical, or microwave, wave (the pump wave) enters a nonlinear optical crystal accompanied by a weak signal whose frequency $\omega_s < \omega_p$. The crystal nonlinearity causes a lossless transfer of power from the pump wave to the signal which is thus amplified. An idler wave, not present at the input, is generated in the process.

The crystal nonlinearity that mediates the power exchange between the pump, idler, and signal is that of the nonlinear polarization oscillating at optical frequencies induced in the crystal by the EM fields. While linear optical phenomena such as loss and dispersion are due to the linear dielectric response,

$$P_i^{\omega} = \chi_{ij} E_j^{\omega},$$

where P_i^{ω} is the complex amplitude of the ith component of the electric polarization induced in a material by a field with amplitude E_j^{ω}. Both the inducing field E_j^{ω} and the induced polarization are at the same frequency ω. χ_{ij}, in general a tensor, is the dielectric susceptibility of the material which can be considered an intrinsic property of the latter. In crystals lacking inversion symmetry, we also expect an induced nonlinear polarization described by a relation between the complex field amplitude at a point and the resulting polarization.

$$P_i^{\omega_3} = d_{ijk} E_j^{\omega_1} E_k^{\pm\omega_2}, \omega_3 = \omega_1 \pm \omega_2. \tag{2}$$

The d_{ijk} coefficients can be derived quantum mechanically, but in the present discussion it is sufficient to consider them classically.

It is a simple matter to ascertain that the nonlinear response of Eq. (2) is the direct equivalent of the voltage-dependent nonlinear capacitor in Heffner's PA [3]. To convert the physics of Eq. (2) to a quantum mechanical parametric interaction, we recall that since a Hamiltonian represents the energy of a system, we need to modify the Hamiltonian of the uncoupled 3-mode model by adding a term accounting for the polarization (P). The dipolar interaction energy (joules/m^3) is $-\bar{P}\cdot\bar{E}$ so that

$$\Delta\mathcal{E}_{\text{interaction}}(\text{joules}) = \int_V d_{ijk} E_i^{\omega_3} E_j^{\omega_1} E_k^{\omega_2} \mathrm{d}^3\vec{r}, \tag{3}$$

where the E's include the modal spatial dependence.

In what follows, we take ω_3 in Eq. (2) as the pump, ω_1 as the signal, and ω_2 as the idler wave. We convert the interaction energy of Eq. (3) to a quantum mechanical operator by quantizing the optical field according to Eq. (4), which describes the quantization of the pump wave. Similar expressions apply to the signal and idler fields. The creation and annihilation operators $a_p^{\dagger}$, a_p, which operate on the EM wave functions and their properties, are treated in any first graduate year text in quantum mechanics. A summary of these relations is in [4].

$$E_p \to \text{const}(a_p^{\dagger} - a_p), \tag{4}$$

$$a_p^{\dagger}|n_p\rangle = \sqrt{n_p+1}|n_p+1\rangle, a_p|n_p\rangle = \sqrt{n_p}|n_p-1\rangle. \tag{5}$$

If we use Eq. (4) and similar equations for the signal and idler in Eq. (3), the result is the quantum mechanical interaction Hamiltonian.

$$\Delta H_I = A(a_p^{\dagger} - a_p)(a_s^{\dagger} - a_s)(a_i^{\dagger} - a_i). \tag{6}$$

This term is added to the unperturbed Hamiltonian to obtain the total Hamiltonian

$$H = \hbar\omega_p(a_p^{\dagger}a_p + 1/2) + \hbar\omega_s(a_s^{\dagger}a_s + 1/2) + \hbar\omega_i(a_i^{\dagger}a_i + 1/2) + A(a_p^{\dagger} - a_p)(a_s^{\dagger} - a_s)(a_i^{\dagger} - a_i).$$

The perturbation ΔH_I can cause transitions between the eigenstates $|n_p\rangle$, $|n_s\rangle$, $|n_i\rangle$ with modal energies $\hbar\omega_p(n_p + 1/2)$, $\hbar\omega_s(n_s + 1/2)$, $\hbar\omega_i(n_i + 1/2)$. Of special interest here is the transition, shown in Fig. 3, from an initial state $|n_p, n_s, n_i\rangle$ to the final state $|n_p - 1, n_s + 1, n_i + 1\rangle$ in which the pump mode loses a quantum of energy $\hbar\omega_p$ while, simultaneously, the signal and idler modes, each, gain a quantum of energy $\hbar\omega_s$, $\hbar\omega_i$, respectively. This transition is due, mathematically, to the term $a_p a_s^{\dagger} a_i^{\dagger}$ in the interaction Hamiltonian (6) when $\omega_p = \omega_s + \omega_i$. The other terms do not conserve energy and thus average out to zero. The rate for this process can be calculated using Fermi's golden rule

$$W_{\text{initial}\to\text{final}} \propto |\langle n_{so}+1, n_{io}+1, n_p-1|a_s^{\dagger}a_i^{\dagger}a_p|n_{so}, n_{io}, n_p\rangle|^2 \times \delta(\omega_p - \omega_s - \omega i) \tag{7a}$$

$$= (n_{so}+1)(n_{io}+1)n_p, \tag{7b}$$

which corresponds to an increase in the initial number of quanta n_{so} and n_{io} of the signal and idler modes. The PA transition between the initial states is indicated in Fig. 3 by the

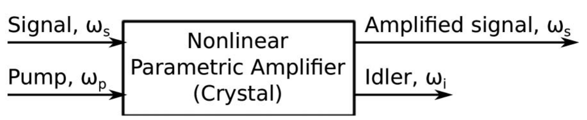

Fig. 2. The basic arrangement for an optical parametric amplifier.

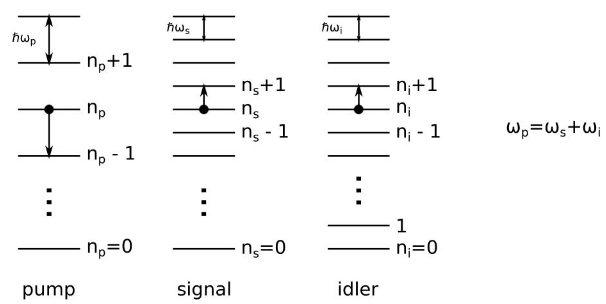

Fig. 3. The quantum states of the system of three modes: pump, signal, and idler. The basic PA process is shown as a quantum transition between pure "harmonic oscillator" states. Transitions originate at the base of the arrows and terminate at the tips.

one-sided arrows. The reverse of this process in which single photons at ω_i and ω_s are annihilated while a pump photon is generated is also possible due to the term $a_s a_i a_p^\dagger$ in Eq. (6).

The PA possesses an input signal (n_{so}) at ω_s while no input exists at the idler frequency ω_i; i.e., $n_{io} = 0$. According to Eq. (7b), the output power at ω_s in this case is proportional to $(n_{so} + 1)n_p$. It is natural to associate the term $n_{so}n_p$ as the amplified signal while considering n_p as the noise, since it corresponds to signal output power not dependent on $n_s 0$.

The description of parametric interactions up to this point was based on transition rates derived from Eq (7a). The treatment is sufficient to point out possible interactions but cannot handle the more subtle issues involving phases and correlations. This requires a more rigorous approach. We solve for the time-dependent annihilation and creation operators in the Heisenberg representation [5]. The resulting equations are

$$\frac{da_s}{dt} = -i\omega_s a_s + i\kappa e^{-i(\omega_p t+\phi)} a_i^\dagger, \frac{da_i^\dagger}{dt} = i\omega_i a_i^\dagger - i\kappa e^{i(\omega_p t+\phi)} a_s, \tag{8}$$

along with their Hermitian adjoints. The solutions to these equations are [5]

$$\begin{aligned} a_s(t) &= \exp(-i\omega_s t)\{a_s(0)\cosh(\kappa t) + ie^{-i\phi}a_i^\dagger(0)\sinh(\kappa t)\}, a_i^\dagger(t) \\ &= \exp(i\omega_i t)\{a_i^\dagger(0)\cosh(\kappa t) - ie^{i\phi}a_s(0)\sinh(\kappa t)\}, \end{aligned} \tag{9}$$

where κ is proportional to the product of χ_{ijk} and the pump field amplitude. It is also proportional to a spatial integral of the products of the three modal fields. The condition that the integral not vanish is the generalized three-dimensional (3D) phase matching condition.

It is interesting to note that although Eqs. (8) and (9) describe the temporal evolution of quantum operators, they are identical in form to the traveling wave (along z) equations of the mode amplitudes in the classical PA [6] if we replace t with z/c. Equation (9) contains all the knowable information of our three-mode quantum system. These equations can be put to work, for example, to describe the case of an optical PA with an input of n_{so} photons and no idler photons. The result is [5]

$$n_s(t) = n_{so}\cosh^2(\kappa t) + (1 + n_{io})\sinh^2(\kappa t) \tag{10a}$$

for the output number of signal quanta $n_s(t)$ and

$$n_i(t) = n_{io}\cosh^2(\kappa t) + (1 + n_{so})\sinh^2(\kappa t) \tag{10b}$$

for the number of idler quanta $n_i(t)$ with initial values of n_{so} and n_{io} photons, respectively. Equation (10a) shows a contribution equal to $\sinh^2(\kappa t)$ to the signal output even with no input signal, $n_{so} = 0$, which agrees with our rate equations approach in Eq. (7b). This term, being independent of n_{so}, represents noise. In the limiting case of high gain, $\kappa t \gg 1$, this noise is equal to the noise power of the laser amplifier discussed above and is equivalent to an effective noise input power of $\hbar\omega_s\Delta\omega_s$.

The quantum analysis of the PA thus settled the issue of which of the two amplifiers, laser or parametric, was superior. The two types of amplifiers had the same basic quantum limit.

An unexpected bonus of the quantum treatment was the prediction of spontaneous generation in a nonlinear optical crystal whereby correlated pairs of signal and idler photons are emitted with only a pump input to the crystal and no signal or idler inputs. This follows directly from Eqs. (10) when we put $n_{io} = 0$, $n_{so} = 0$, i.e., no input signal and idler fields. The only incident optical field on the crystal is that of the pump wave represented here by the coupling constant κ. In this case

$$n_s(t) = n_i(t) = \sinh^2(\kappa t) \tag{11}$$

so that equal numbers of signal and idler photons are generated without corresponding inputs. This phenomenon became known as spontaneous parametric fluorescence and was observed later by Harris *et al.* [7].

Another area of investigation closely related to the subject at hand and which has been growing in importance recently is that of the interaction of light and hypersound. This field can trace its beginning to the observation of stimulated Brillouin scattering in 1964 by Chiao *et al.* [8]. As it turns out, the quantum theory for 3-wave interactions described above can be applied, formally, to the case of acoustic waves-optical waves interactions [9].

One can simply replace the signal wave, or the idler, of the optical parametric interaction with a hypersonic wave or mode. By quantizing the hypersonic wave [10] and introducing the corresponding phonon creation and annihilation operators, we obtain an interaction Hamiltonian

$$H_I = \kappa(a_s^\dagger - a_s)(a_p^\dagger - a_p)(a_i^\dagger - a_i) + \text{hermitian adjoint} \tag{12}$$

where $a_s^\dagger$ and a_s are the creation and annihilation operators for phonons and $a_i^\dagger$, a_i, $a_p^\dagger$, a_p refer to the EM idler and pump waves. The coupling constant κ is proportional to the electrostrictive coefficient of the medium (or, equivalently, the photoelastic coefficient). The formal analogy between Eqs. (12) and (6) results in a direct transference of all the optical parametric phenomena to the optical-sound wave case. The most interesting of these is the parametric sound optical oscillation, in which optical pumping at ω_p above a threshold value results in a simultaneous generation of a phonon at ω_s and a photon at $\omega_i = \omega_p - \omega_s$, while a photon at ω_p is annihilated. In a manner completely analogous to the all-optical PA, this process is due to the term $\kappa a_s^\dagger a_i^\dagger a_p$ in Eq. (12), which plays the same role as a similar term in Eq. (7a), except that here the operators a_s and $a_s^\dagger$ operate on the hypersound mode. Recently, a number of independent demonstrations of hypersound-optical oscillators has been reported [11]. The generation of a phase-correlated pair of a phonon at ω_s and a photon at ω_i, i.e., no threshold, must have its spontaneous emission equivalent in which the correlated photon-phonon pairs are omitted spontaneously without a threshold. The observation of this phenomenon will require a cooling of the hypersound medium (resonator) to a temperature $T < \hbar\omega_s/k \approx 10$ K for $\omega_s/(2\pi) \approx 3 \times 10^{10}$ Hz.

The above constitutes a personal account of how an attempt to answer some pragmatic engineering questions led to the formulation of quantum nonlinear optics and to the theoretical foundation of a number of new phenomena and devices.

REFERENCES

1. A. L. Schawlow, and C. H. Townes, “Infrared and optical masers,” Phys. Rev. **112**, 1940–1949 (1958).
2. R. Serber, and C. H. Townes, “Limits on electromagnetic amplification due to complementarity,” in *Quantum Electronics*, C. H., Townes, ed. (Columbia University Press, 1960), pp. 233–255.
3. H. Heffner, “Parametric amplifiers and their comparison with masers,” in *Quantum Electronics*, C. H., Townes, ed (Columbia University Press, 1960), p. 269.
4. See, for example, A. Yariv, *Quantum Electronics* (John Wiley and Sons, 1986), pp. 54–56.
5. W. H. Louisell, A. Yariv, and A. E. Siegman, “Quantum fluctuations and noise in parametric processes,” Phys. Rev. **124**, 1646–1654 (1961).
6. A. Yariv, and P. Yeh, *Photonics*, 6th ed. (Oxford University Press, 2007), pp. 370–371.
7. S. E. Harris, M. K. Oshman, and R. L. Byer, “Observation of tunable optical parametric fluorescence,” Phys. Rev. Lett. **18** 732–734 (1967).
8. R. Y. Chiao, C. H. Townes, and B. P. Stoicheff, “Stimulated Brillouin scattering and coherent generation of intense hypersonic waves,” Phys. Rev. Lett. **12** 592–595 (1964).
9. A. Yariv, “Quantum theory for parametric interactions of light and hypersound,” IEEE J. Quantum Electron. **1** 28–36 (1965).
10. C. Kittel, *Quantum Theory of Solids* (John Wiley and Sons, 1963).
11. J. B. Khurgin, “Phonon lasers gain a sound foundation,” Physics **3**, 1–3 (2010).